

21:48 04277108//

2004年2月10日(火)16:44/審査16:43/文書番号4801505004 B2,4,

FEB 27 2004

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In Re Application Of:

Masaki Fujiwara Et Al.

Serial No: 09/689,817

Filed: October 13, 2000

: Group Art Unit: 1745

Examiner: Tsang Foster, Susy

FOR: MOLDED ELECTRODE, METHOD FOR PRODUCTION THEREOF, AND  
SECONDARY BATTERY USING THEREOF

DECLARATION UNDER 37 CFR 1.132

Honorable Commissioner of Patents and Trademarks  
Alexandria, Virginia 22313-1450

Sir:

I, Tetsuya YOSHINARI, am an employee of NEC TOKIN Corporation  
which is the present assignee of the above-identified application.

1. I was born on June 12, 1974. I graduated from Okayama University of  
Science in March 1997 and received a master degree in Science at Himeji Institute  
of Technology in March 1999.

2. I had been employed by NEC TOKIN Corporation since April 1, 1999  
and working at the Research & Development Unit of NEC TOKIN Corporation. I  
have worked in the battery field for one year and 7 months.

3. The following experiments were carried out by me or persons under my  
supervision and control in order to clarify the advantages of the molded electrode  
according to the present invention:

## EXPERIMENTS

### *Example A-1 to A-4*

In Examples A-1 to A-4, batteries were formed according to the method described in Example 6 of the present specification except the pressures for molding positive electrodes were selected as shown in Table 1. Particulars are summarized as follows.

#### (1) Material

##### (i) Positive electrode

Active material: Polycyanoindole doped with sulfate ion (80% by weight)

Conductivity-enhancing agent: Vapor phase-grown carbon (10% by weight)

Plasticizer: Butyl butylphthalylglycolate (10% by weight)

Current collector: Tantalum foil 50  $\mu$ m

##### (ii) Negative electrode

Active material: Polyphenylquinoxaline (75% by weight)

Conductivity-enhancing agent: Vapor phase-grown carbon (15% by weight)

Plasticizer: Butyl butylphthalylglycolate (10% by weight)

Current collector: Tantalum foil 50  $\mu$ m

##### (iii) Electrolytic solution

aqueous solution of 2 M of sulfuric acid

#### (2) Preparation of electrodes

##### (i) Positive electrode

Polycyanoindole doped with sulfate ion was prepared in the same manner described in the present specification on page 36, lines 11 to 19 in Example 3. In the similar manner described in Example 1 of the present invention, the obtained powder of doped polycyanoindole was mixed thoroughly with vapor phase-grown carbon and butyl butylphthalylglycolate, and the mixture was molded into 3 mm thickness on a tantalum foil by pressure described in Table 1 at 200 °C for 30 minutes.

##### (ii) Negative electrode

As described in the present specification on page 43, line 14 to page 44, line 5, Polyphenylquinoxaline powder, a vapor phase-grown carbon and butyl

butylphthalylglycolate were mixed thoroughly, and the mixture was molded into 3 mm thickness on a tantalum foil by pressure of 150 kgf/cm<sup>2</sup> at 200 °C for 30 minutes to give negative electrode of density of 0.84 g/cm<sup>3</sup>.

#### *Example B-1 and B-2 (Comparative Example)*

In Examples B-1, batteries were formed according to the method described in Comparative Example 5 of the present specification. In Examples B-2, 1 % of dioctyl phthalate was further added to the electrode materials. Particulars are summarized as follows.

##### **(1) Material**

###### **(i) Positive electrode**

Active material: Polycyanoindole doped with sulfate ion (80 % by weight)

Conductivity-enhancing agent: Vapor phase-grown carbon (10 % by weight)

Binder: Polyvinylidene fluoride (10% by weight)

Plasticizer (Example B-2 only): Dioctyl phthalate (1 % by weight to the above)

Current collector: Nickel current collector

###### **(ii) Negative electrode**

Active material: Polyphenylquinoxaline (65% by weight)

Conductivity-enhancing agent: Vapor phase-grown carbon (35% by weight)

Current collector: conductive rubber

###### **(iii) Electrolytic solution**

aqueous solution of 2 M of sulfuric acid

##### **(2) Preparation of electrodes**

The above materials were mixed in appropriate amount of N-methyl 2-pyrrolidone and coated on a nickel current collector in case of formation of positive electrode and on a conductive rubber in case of formation of negative electrode by a doctor blade, followed by drying at 120 °C for 15 minutes. The resultant positive electrode and negative electrode had thickness of 500  $\mu$  m.

## **Results**

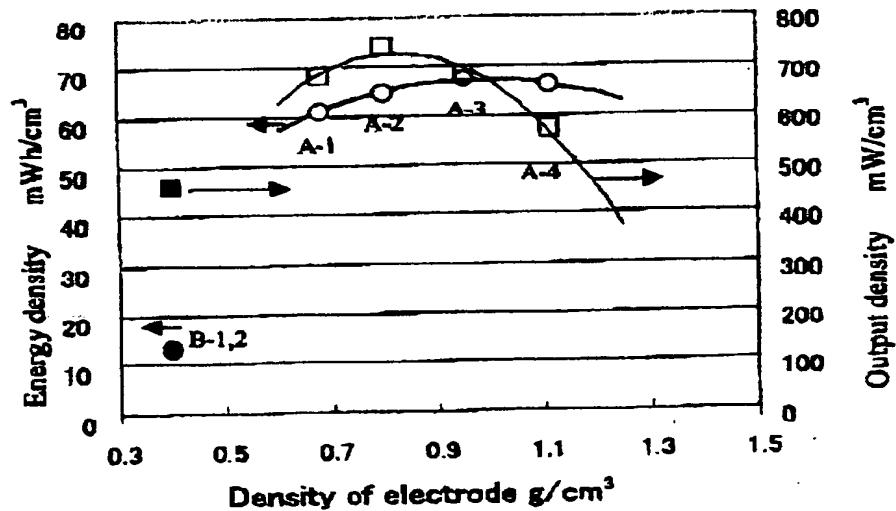
Densities of the electrodes were given in Table 1. Energy density was measured as defined in the present specification and Output density was calculated by (voltage at just after discharge) x (discharge current when reduction ratio of capacity becomes 50 %)

As shown in Table 1 and Fig. 1, the result clearly shows that Example B-1 and B-2 where the electrodes were made by conventional solvent-coating method do not show the sufficient energy density and output density. Whereas, Examples A-1 to A-4 where the molded electrodes were used shows excellent energy density and output density. In Example B-2, 1% of dioctyl phthalate as a plasticizer was contained in the positive electrode. The amount of the plasticizer was chosen so as to match the amount used in example 2 of JP08-064200. However, even if the plasticizer is contained, non-molded electrode never gives good battery properties as shown by Example B-2. The difference of the battery properties is attributed to the difference in density of electrode. Thus, the electrode obtained by molding is different from that obtained by solvent-coating method as product itself.

Table 1

	Press- pressure (kgf/cm <sup>2</sup> )	Density of electrode (g/cm <sup>3</sup> )	Energy density (mWh/cm <sup>3</sup> )	Output density (mW/cm <sup>3</sup> )
Example A-1	25	0.68	60.9	678
Example A-2	75	0.8	64.6	740
Example A-3	125	0.9	67.1	680
Example A-4	250	1.1	66.0	572
Example B-1	Solvent- coating	0.4	13.1	459
Example B-2	Solvent- coating	0.4	13.0	450

Fig. 1



I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardized the validity of the above application or any patent issued thereon.

February 6, 2004.  
(Date)

Tetsuya Yoshinari  
Tetsuya YOSHINARI